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Earth and Planetary Science Letters 171 (1999) 325–328

EPSL

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Express Letter

Dependence of the decay rate of ^7Be on chemical forms

Chih-An Huh*

Institute of Earth Sciences, Academia Sinica, Taipei, Taiwan, ROC

Received 25 May 1999; revised version received 6 July 1999; accepted 6 July 1999

Abstract

The decay rate of ^7Be depends on the electron density at the nucleus and therefore may vary with the chemical environments of the Be atom. Using a 100% efficiency high-purity germanium detector, the decay constants of ^7Be in $\text{Be}^{2+}(\text{OH}_2)_4$, $\text{Be}(\text{OH})_2$ and BeO have been measured at unprecedented high precision (of $\pm 0.01\%$), resulting in $T_{1/2} = 53.69$ d, 53.42 d, and 54.23 d, respectively. The observed difference, by as much as 1.5%, clearly indicates that decay rates of nuclides undergoing electron capture decay are not necessarily constant as has always been assumed in geological, oceanographic, and environmental studies. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Be-7; radioactive decay; absolute ages; gamma-ray spectroscopy

1. Introduction

Beryllium-7 is the lightest radioactive nuclide that decays by capturing an orbital electron from the innermost electron shell (*1s* or *K*-shell) at its nucleus. It was first proposed more than a half century ago that the decay rate of ^7Be could be altered by changing the electron density at the nucleus [1,2]. Following this suggestion, numerous attempts were made to determine the variations of the decay rate of ^7Be in various chemical forms [3–12]. However, these experiments were primarily performed 4 to 5 decades ago [3–11] and the most recent report was published in 1970 [12]. During that time period, the detection of ^7Be decay was conducted using ionization chambers or NaI detectors. The reported errors for the decay rate associated with such mea-

surement techniques were generally of the order of 0.1–1%, making it rather difficult to resolve the subtle difference of the ^7Be decay rate among different combinations of Be and anions. Considering the advances in semiconductor technologies and the lack of new measurements in the past three decades, it is expedient to revisit this issue now. Using an advanced HPGe γ -spectrometry system, the decay constants of ^7Be in $\text{Be}^{2+}(\text{OH}_2)_4$, $\text{Be}(\text{OH})_2$ and BeO have been measured at unprecedented high precision. The experimental procedures and results are reported below.

2. Methods and results

Carrier-free ^7Be (in 1 ml of 0.1 N HCl) with an activity of 1.1 mCi as of November 9, 1998 was obtained from the Hot Lab of Brookhaven National Laboratory (S/M number: 054911). From the

* Tel.: +886-2-2783-9910/ext. 607; Fax: +886-2-2783-9871; E-mail: huh@earth.sinica.edu.tw

^7Be stock solution, counting sources in three different chemical forms were prepared. The stock solution itself provided one chemical state for Be, i.e., $\text{Be}^{2+}(\text{OH}_2)_4$, with the Be^{2+} ion surrounded by four water molecules in the solution. The $\text{Be}^{2+}(\text{OH}_2)_4$ counting source was prepared by directly transferring 5 μl of the stock solution into a KimaxTM counting tube. With an inner diameter of 10 mm and an outer diameter of 13 mm, the tube fits ideally into the well of the HPGe detector for gamma counting.

The $\text{Be}(\text{OH})_2$ and BeO sources were prepared from 100 μl of the stock solution by adding 5 mg of ^9Be (initially dissolved in 1 ml of 1 N HCl) as the Be carrier followed by NH_4OH to form $\text{Be}(\text{OH})_2$ precipitate. After centrifugation to separate the precipitate from the solution, the supernatant water was removed. About one-half of the precipitate was transferred to a counting tube and used as the $\text{Be}(\text{OH})_2$ counting source. The remaining half was transferred to a platinum crucible and heated in a muffle furnace at 1000°C for 1 h to convert $\text{Be}(\text{OH})_2$ into BeO. The oxide powder was then transferred to a counting tube and used as the BeO counting source.

The HPGe well detector (EG&G ORTEC GWL-100230) used in this experiment is interfaced with a digital γ -ray spectrometer (EG&G ORTEC DSPecTM). The counting system provides extended live-time correction according to the Gedcke–Hale method [13] and automatically sets a threshold to reject pile-up of pulse-pair at 0.5 μs resolution. The detector has an active volume of 100 cm^3 , a well diameter of 1.45 cm and a well depth of 4 cm. It offers a counting efficiency of 100% (relative to 3×3 NaI) and a resolution of 1.45 keV (FWHM) for the ^7Be photon peak centred at 477.56 keV. With the counting tubes placed in the detector well, the Be sources were very close to the bottom of the well, providing an absolute counting efficiency of $\sim 21\%$ for the ^7Be peak. A typical spectrum obtained from the $\text{Be}^{2+}(\text{OH}_2)_4$ source is shown in Fig. 1. Besides the ^7Be peak, only a small ^{54}Mn peak (at 834.81 keV) appeared in the spectrum. Activity of the ^{54}Mn impurity contained in the ^7Be stock solution was 0.28 $\mu\text{Ci/ml}$, or about 0.025% of the ^7Be activity at the beginning of this experiment. It is impossible for the ^{54}Mn impurity to interfere with the counting of ^7Be . The ^{54}Mn peak was not seen in the spectra obtained from the $\text{Be}(\text{OH})_2$ and the BeO sources, in-

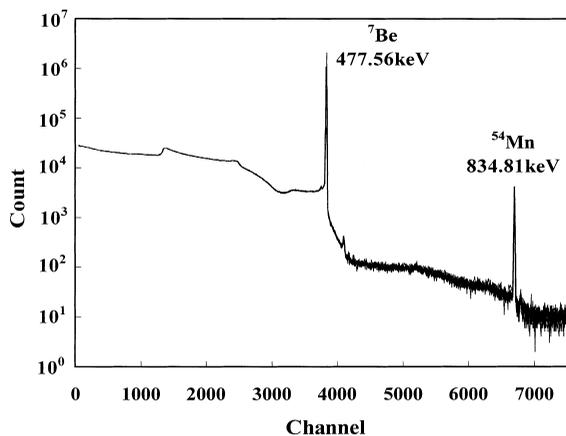


Fig. 1. Typical γ -ray spectrum from the decay of ^7Be in the $\text{Be}^{2+}(\text{OH}_2)_4$ source. Besides the ^7Be peak, a small ^{54}Mn peak (at 834.81 keV) appeared in the spectrum.

dicating effective removal of Mn during the chemical procedures.

Activities of ^7Be in the three sources were assayed on a daily basis (or as frequent as possible) and lasted 70–120 days. To compensate for the decrease of count rate with time and maintain compatible counting statistics between runs, the counting time increased from <20 min at the beginning to >1 h near the end of the experiment. The accumulated counts from ^7Be decay for each run were generally on the order of 10^6 – 10^7 , resulting in typical counting errors of ~ 0.03 – 0.1% ($\pm 1\sigma$) for the data points. The stability of the system was ensured by weekly calibration of the detector using a uraninite standard solution and a ^{152}Eu source whose activity was comparable to that of the ^7Be sources. Throughout the course of this experiment there were no signs of any changes in energy, resolution and efficiency.

The exponential decrease of ^7Be activity (A_t) with time in the three sources is shown in Fig. 2. To simplify the plots, A_t is normalized against A_0 (^7Be activity at $t = 0$). Weighted least-squares fitting of the data sets results in decay constants ($\pm 1\sigma$ uncertainties) of 0.0129092 ± 0.0000014 , 0.0129765 ± 0.0000014 and 0.0127825 ± 0.0000014 d^{-1} (or half-lives of 53.694 ± 0.006 , 53.416 ± 0.006 and 54.226 ± 0.006 d) for ^7Be in $\text{Be}^{2+}(\text{OH}_2)_4$, $\text{Be}(\text{OH})_2$, and BeO, respectively. It is important to note that the $\sim 1.5\%$ difference between the decay

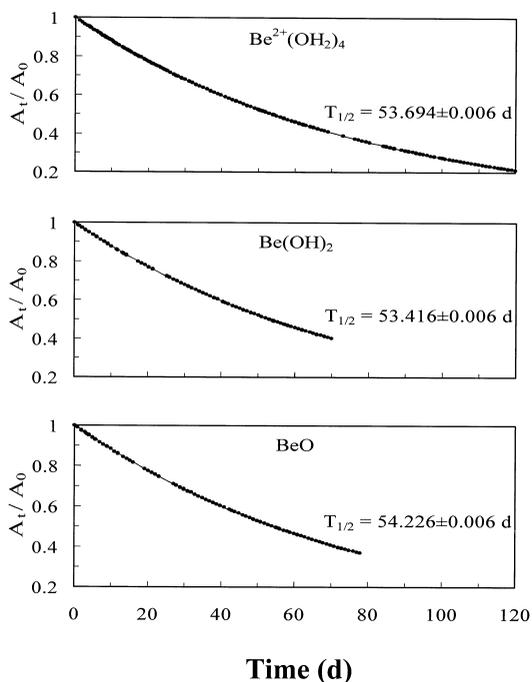


Fig. 2. Exponential decay of ${}^7\text{Be}$ in (from top to bottom) the $\text{Be}^{2+}(\text{OH}_2)_4$, $\text{Be}(\text{OH})_2$ and BeO sources. Error bars for the A_t/A_0 values are actually much less than the size of the data points.

constants for ${}^7\text{Be}$ in $\text{Be}(\text{OH})_2$ and BeO is one order of magnitude larger than the reported difference of $\sim 0.1\text{--}0.2\%$ in the decay constants of ${}^7\text{Be}$ among other chemical forms of Be [12].

The decay rate of ${}^7\text{Be}$ in the chemical form of $\text{Be}(\text{OH})_2$ is reported here for the first time, and its validity can be tested rigorously. In a concurrent experiment dealing with pressure dependence of the decay rate of ${}^7\text{Be}$ [14], $\text{Be}(\text{OH})_2$ was used as the carrier phase of ${}^7\text{Be}$ as well as the pressure medium. In order to accommodate the geometry and weight of the pressure cells, a different detector (ORTEC EG&G GEM-150230) was used. The control sample (i.e., at 1 atm) for the pressure experiment was prepared by directly mounting the $\text{Be}(\text{OH})_2$ source on a glass slide. The decay rate of ${}^7\text{Be}$ in uncompressed $\text{Be}(\text{OH})_2$ obtained from this independent experiment, under a different detector–sample configuration, is identical to the value reported here, lending unequivocal support to the accuracy and precision of our measurements.

3. Discussion

The decay half-life of ${}^7\text{Be}$ reported previously in the literature falls in the range 52.93–53.61 d ([15] and references therein), and the weighted average of 53.3 d is generally adopted in various applications using this nuclide. The uncertainties of these measurements vary from less than 0.2% (e.g., [13,16,17]) to greater than 0.5%. Therefore, error bars of these measurements do not always overlap. Because the chemical forms of Be in these measurements were often unspecified, it is not clear whether the difference is due to different chemical environments or is simply caused by experimental errors. In the present study, by measuring the decay rate of ${}^7\text{Be}$ in three common forms of Be with an unprecedented high precision of $\sim 0.01\%$, it was shown that the half-life of ${}^7\text{Be}$ in natural environments could vary by as much as 1.5%. The variation can be explained by a change in electron density around the nucleus of Be atom due to its association with different anions, and hence different electronic polarizability and dipole moments. Besides ${}^7\text{Be}$, some other nuclides having important geochemical applications (e.g., ${}^{26}\text{Al}$, ${}^{36}\text{Cl}$, ${}^{40}\text{K}$, etc.) also undergo electron capture decay. Thus, decay rates of these nuclides may also depend on their chemical forms, but the effect will probably be smaller for heavier nuclides due to a better shield of K -shell electrons by more electrons and shells.

Acknowledgements

The author gratefully acknowledges the assistance of Dr. Jason Shen in the acquisition of the ${}^7\text{Be}$ source and Drs. Tibor Dunai and Doug Hammond for critical review of the manuscript. This work was supported by the National Science Council and the Atomic Energy Commission. [RV]

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